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9.1 INVESTIGATION OF PHOTOCHEMICAL MODELING OF POINT SOURCE POLLUTANTS WITH EULERIAN GRID AND LAGRANGIAN PLUME APPROACHES

James M. Godowitch*

Atmospheric Sciences Modeling Division
Air Resources Laboratory
National Oceanic and Atmospheric Administration
Research Triangle Park, North Carolina

1. INTRODUCTION

Significant amounts of nitrogen oxides (NOx) and sulfur oxides (SOx) are emitted from individual elevated point sources. The primary emission of NOx, in particular, are important precursors in the photochemical generation of a variety of secondary oxidants, such as ozone (O_3). Since major point sources are distributed inhomogeneously in space and the physical dimensions of a pollutant plume increase at a finite rate with distance downwind, a key aspect for realistic modeling of this notable class of emissions is to be able to properly resolve the spatial scale of the pollutant plume. From a photochemical perspective, a fresh power plant plume represents a high NOx, low HC (hydrocarbon) regime, while the surrounding environment can often be in the exact opposite regime. As the plume spreads horizontally and vertically due to atmospheric dispersion processes, it evolves chemically as background pollutants are entrained and mixed with in-plume pollutants. Consequently, model resolution and plume growth rate are important factors which strongly influence the photochemical evolution of plumes.

The modeling approaches selected to simulate anthropogenic emissions include the Eulerian grid and Lagrangian plume techniques. The widely-used treatment in many Eulerian grid models is to inject the point source emissions into a particular grid cell. However, the initial lateral dimension of a plume is generally much smaller than the grid cell size. Thus, the instant dilution of the primary emissions of the pollutants over the entire grid cell volume can strongly distort the non-linear photochemistry process and contribute to model uncertainty. To compensate for overdilution of a plume, a smaller grid cell size can be specified. In contrast, the Lagrangian plume model approach features a set of expanding, contiguous cells representing the overall crosswind dimension of a plume. Each plume cell is treated as a single vertical column extending over the mixing layer, while an Eulerian grid model generally contains multiple vertical layers within the boundary layer. Nevertheless, the artificial dilution due to dumping of the point emissions into a grid cell is more severe in the horizontal.

In this paper, results of Eulerian grid and Lagrangian photochemical model simulations of emissions from a major elevated point source are presented. A series of simulations with grid sizes varying from 30 km to 2 km were performed with the Urban Airshed Model, a photochemical grid model, in order to examine the capability of the model to resolve plume features and to emulate the chemical evolution of the pollutant plume. Differences in pollutant concentrations with emphasis on O_3 for various grid sizes are highlighted. The grid model simulation results are compared to O_3 concentrations generated from a Lagrangian reactive plume model. Both models were applied to treat the transport, dispersion, and photochemical processes impacting a pollutant plume. In order to promote the comparison and interpretation of the model results, meteorological inputs for the Lagrangian plume model were provided through an interface program using the input data sets employed to drive the grid model.

2. MODEL DESCRIPTION

2.1 Eulerian Grid Model

The Urban Airshed Model (UAM) is a three-dimensional, Eulerian photochemical grid model that treats the physical and chemical processes affecting pollutant concentrations from both surface area and/or elevated point sources. The regulatory model version (UAM-IV), described by Scheffe and Morris (1993)

* On assignment to the National Exposure Research Laboratory, U.S. Environmental Protection Agency

Author Address: J. Godowitch, US EPA, MD-80, Research Triangle Park, NC 27711

has been widely applied in metropolitan areas required to demonstrate attainment of the maximim hourly O_3 standard. The UAM is based on the atmospheric continuity equation for a pollutant species, which represents the mass balance of emissions, transport, diffusion, photochemical reactions, and dry deposition processes. Of particular relevance, its treatment for turbulent diffusion involves a constant horizontal eddy diffusivity coefficient (K_y= 50m^2/s). However, a significant factor contributing to horizontal dispersion in grid models comes from the numerical advection scheme. In the standard UAM, the Smolarkiewicz (1983) method is used as the horizontal advection technique. A vertical eddy diffusivity (K_z), which is a function of stability and turbulence parameters, maintains strong vertical exchange between model layers below the mixing height. Finally, the Carbon Bond IV mechanism (CBM-IV) containing 86 reactions and 33 individual species simulates the photochemical reaction processes. Emissions from a point source are processed to determine plume to determine the appropriate vertical layer for injection. Morris and Myers (1990) also give additional details about the techniques for the atmospheric processes in the regulatory UAM and running the modeling system.

In this modeling study, a modified version of the UAM was applied. Modifications were made to allow for 3-D inputs of additional meteorological parameters as described by Godowitch et al. (1992). In addition, the horizontal advection scheme of Bott (1989), which exhibits less numerical diffusion (Odman et al., 1995), was installed as an alternative to the method in the regulatory UAM.

2.2 Lagrangian Reactive Plume Model

The reactive plume model version IV (RPM-IV) described in EPA (1993) is a Lagrangian photochemical plume model that simulates the transport, diffusion, and chemistry in an array of adjacent cells representing a plume cross-section perpendicular to the wind flow. The RPM-IV model treats the entrainment of ambient pollutants into the edge plume cells and entrainment/detrainment between inner plume cells gradually as the horizontal dimension of the plume increases downwind. The transport speed, and the horizontal and vertical dimensions of the plume section must be provided as a function of downwind distance. The model was designed to simulate plume sections under convective conditions since each plume cell represents a single well-mixed vertical column over the depth of the mixing layer. The plume model employs the same CBM-IV photochemical mechanism as the UAM. A more detailed description of the development and attributes of this model are provided by Stewart and Liu (1981).

3. MODEL SIMULATIONS AND RESULTS

Both models were applied to simulate the concentrations in the plume emanating from a major elevated point source situated west of St. Louis, Missouri for a historical case study day (9 July 1976) when experimental measurements were also obtained to characterize the transport, dispersion, and transformation of the pollutant plume during the daytime (Gillani and Wilson, 1980). The emission rate of 1.88 kg/s of NOx for each model was prescribed with NO representing 95% of the emissions and the remainder as NO_2. The stack parameter information consisted of a 214 m stack height, 8.8 m stack diameter, 27.4 m/s exist velocity, and a 441 °K exit temperature.

A modeling domain was defined to be 240 km on a side and a series of simulations were performed with grid cell sizes of 2, 4, 8, 16, and 30 km. While the latter grid specifications are comparable to typical regional scale grid cells, the 2 km grid cell size is close to a lower limit for the grid model. The vertical model layer structure, except for the 2 km grid size, was typical of most applications with 5 layers; 2 layers below and 3 layers above the mixing height. For the 2 km grid simulation, one lower and one upper layer were modeled. Gridded input data sets for each cell size were generated by a meteorological processor program (Godowitch et al., 1992), which employs diagnostic/interpolative methods to derive the hourly 3-D wind, temperature, and the 2-D mixing height fields. Observations included the routine surface hourly observations from Lambert STL airport and upper air profiles obtained four times daily at four special-study rawinsonde sites in the region. Detailed pollutants measurements needed to define initial and boundary conditions were unavailable. Since this study is not intended as a model evaluation effort, the same initial (IC) and constant boundary (BC) conditions were specified with reasonable background values for consistency in all simulations. Concentrations levels for IC and BC included; total HC of 100 ppbC partitioned into the various organic classes defined in the CBM mechanism, NO set to 0.25 ppb, NO_2 at 0.75 ppb, and O_3 set at 60 ppb. Model runs were performed with point source emissions starting at 12 LST with

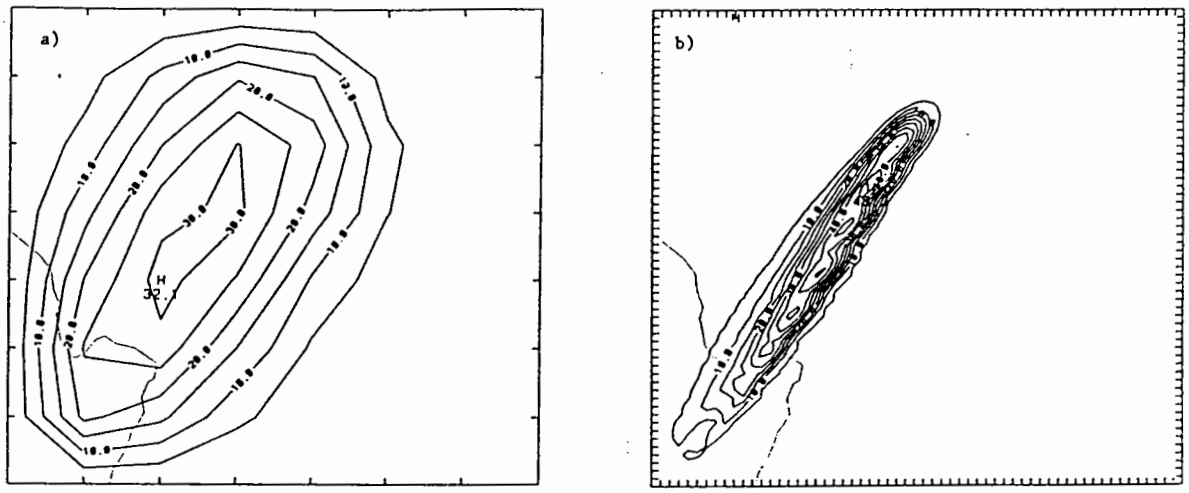

Figure 1. Spatial patterns of maximum excess ozone above background of the point source plume from 12-18 LST from grid model simulations at grid cell sizes of a) 30 km and b) 4 km with the same domain. The Mississippi River (dotted line) is also displayed.

continuous emission rates for six hours. Surface area emissions were prescribed to be zero in order to focus on the modeled plume concentrations.

The maximum excess O_3 field above background over the afternoon period is displayed in Figure 1 to illustrate the differences in the overall size of the pollutant pattern and O_3 concentration in the plume for a coarse (30 km) and a fine (4 km) grid simulation. The 30 km grid results exhibit a considerably broader plume O_3 pattern and the excess plume ozone above background is less than in the 4 km grid size results. The coarse grid cell size provides for much greater dilution of the primary NO emissions. Of relevance to the photochemical process, the HC/NOx ratio in the source cell for these grid sizes differed by nearly a factor of 10 with values near 30 for the coarse grid simulation. Consequently, photochemical O_3 production started in the source cell and the peak O_3 occurred relatively close to the source for this coarse grid. Table 1 contains results from the model simulations for the different grid sizes. It reveals that as the grid size decreased the downwind location of the maximum O_3 concentration in the plume increased. Another interesting result in Table 1 is that a notable decreasing trend in the excess O_3 occurred at the finer grid sizes of 2 and 4 km. These grid cell sizes are much closer to actual plume dimensions, especially at an early stage.

The O_3 values relative to background for the plume at hour 18 LST from the 2 km grid simulation are shown in Figure 2. It reveals that important features of chemical evolution in the plume as described by Gillani and Wilson (1980) were captured. Up to about 50 km downwind an ozone deficit below background exists in the plume due to ozone depletion by high NO concentrations from the primary emissions. Ozone concentrations recover gradually in the plume's core and in an intermediate phase, O_3 formation along the plume edges occurs as mixing with higher HC concentrations contained in surrounding grid cells leads to characteristic bulges in O_3 concentrations. Further downwind for this source strength, the maximum excess O_3 in the plume is found in the mature chemical phase.

For simulations with the Lagrangian RPM, inputs for winds, mixing heights, temperatures, and background concentrations were obtained from the appropriate grid cell where the plume cross-section was situated by applying an interface processor designed to couple these models (EPA, 1993). An additional grid model simulation was conducted with zero emissions in order to generate gridded background concentrations for use with the plume model. Furthermore, lateral plume dimensions were provided from an analytical curve fit to observed plume widths (Godowitch et al., 1995). The RPM simulation commenced at 12 LST with the release of a single plume cross-section using the same emission rates as the grid model and the plume section was modeled for 8 hours.

Table 1. Grid Model Results from Point Source Emissions Simulations

Grid Cell Size (km)	Excess Plume Ozone* (ppb)	Downwind Distance (km)
30	32.1	67
16	43.6	71
8	49.4	102
4	46.0	101
2	40.0	112

* Note: Concentration values minus background for hour 17-18 LST

Figure 3 depicts the O_3 values relative to background concentration for the expanding horizontal plume section at various downwind distances. The notable features of the evolutionary cycle of plume O_3 are certainly evident in these selected plume cross-sections. A deep ozone deficit occurs near the source, and the transition toward a recovery of the O_3 concentrations to near background values is seen in the plume section at 50 km downwind, as well as characteristic bulges of ozone along each edge of the plume. A maximum excess ozone of about 25 ppb was produced in the 150 km downwind distance displayed in Figure 3. The maximum O_3 generated in the RPM was less when compared to the grid model values, even in the 2 km grid simulation. A preliminary explanation for the differences in the excess O_3 between these modeling approaches is their different treatments and rates of diffusion, however, further analyses are warranted.

4. SUMMARY

A limited modeling study of point source emissions was performed with Eulerian grid and Lagrangian plume models to examine the comparative predictions of ozone concentrations in a pollutant plume governed by transport, diffusion, and photochemical processes. Results indicated that the grid model was able to replicate the evolution of ozone in the pollutant plume for this rather large emissions source at the finest grid resolution. As anticipated, simulation results for different grid cell sizes also revealed the maximum O_3 occurred further downwind as the model grid size was decreased, and the peak ozone level was lowest for the coarsest grid size (30 km). Interestingly, maximum O_3 values were on a downward trend for the selected smaller grid cell sizes for the large emissions source strength employed in this study. A comparison between the modeling approaches indicated the maximum ozone generated from the RPM approach was lower than values from the grid model simulations. Since attempts were made to simulate the same processes and provide common inputs to both models, the difference in excess plume ozone is primarily attributed to the different technical methods for diffusion in these modeling approaches. Additional analysis of the results to assess the impact on other secondary species concentrations (ex., PAN, HNO_3) and model runs with other (lower) emission rates are underway.

Considerable interest exists and efforts are underway in plume-in-grid treatments, whereby a plume module is embedded in an Eulerian grid framework to treat point source plumes during a subgrid scale phase. The plume-in-grid approach could greatly reduce the rather high computational burden encountered with the fine scale gridded domains used in this study. Further modeling studies of each modeling approach, such as this one, are warranted since results could have important implications for plume-in-grid treatments and can also provide valuable information for the transfer of pollutant plumes over to the grid system.

Disclaimer - This paper has been reviewed in accordance with the U.S. Environmental Agency's peer and administrative review policies and approved for presentation and publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

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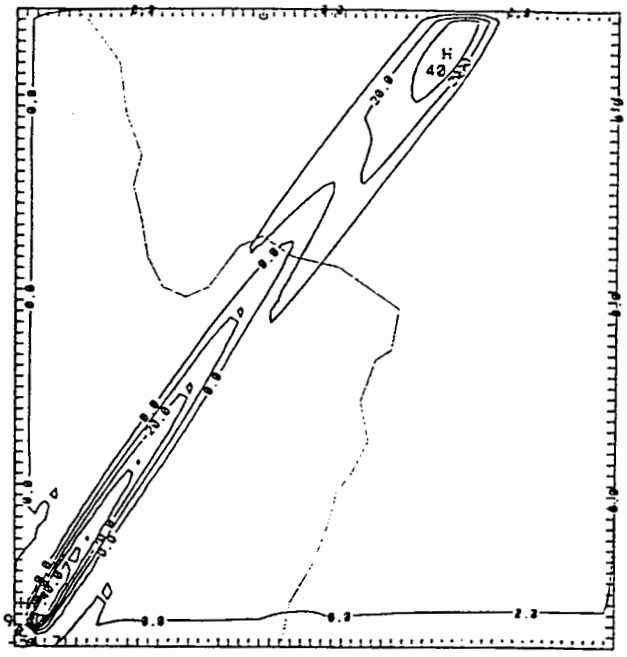

Figure 2. Relative ozone values with respect to background in the pollutant plume from the grid model simulation with a 2 km grid resolution at hour 18 LST. The Mississippi River (dotted line) is also displayed.

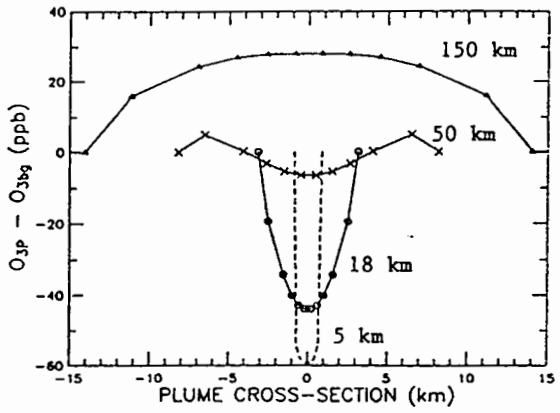

Figure 3. Relative ozone values in the horizontal plume section from the RPM simulation at various downwind distances depict the chemical evolution of ozone in the point source plume. The RPM simulated 10 subplume cells, with the value on each edge being the background ozone specified by the grid model.

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15. SUPPLEMENTARY NOTES

16. ABSTRACT

In this paper, results of Eulerian grid and Lagrangian photochemical model simulations of emissions from a major elevated point source are presented. A series of simulations with grid sizes varying from 30 km to 2 km were performed with the Urban Airshed Model, a photochemical grid model, in order to examine the capability of the model to resolve plume features and to emulate the chemical evolution of the pollutant plume. Differences in pollutant concentrations with emphasis on O_3 for various grid sizes are highlighted. The grid model simulation results are compared to O_3 concentrations generated from a Lagrangian reactive plume model (RPM). Both models were applied to treat the transport, dispersion, and photochemical processes impacting a pollutant plume. In order to promote the comparison and interpretation of the model results, meteorological inputs for the Lagrangian plume model were provided through an interface program using the input data sets employed to drive the grid model. Results indicated that the grid model was able to replicate the evolution of ozone in the pollutant plume for this rather large emissions source at the finest grid resolution. As anticipated, simulation results for different grid cell sizes also revealed the maximum O_3 occurred further downwind as the model grid size was decreased, and the peak ozone level was lowest for the coarsest grid size (30 km). Interestingly, maximum O_3 values were on a downward trend for the selected smaller grid cell sizes for the large emissions source strength employed in this study. A comparison between the modeling approaches indicated the maximum ozone generated from the RPM approach was lower than peak values from the grid model simulations.

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